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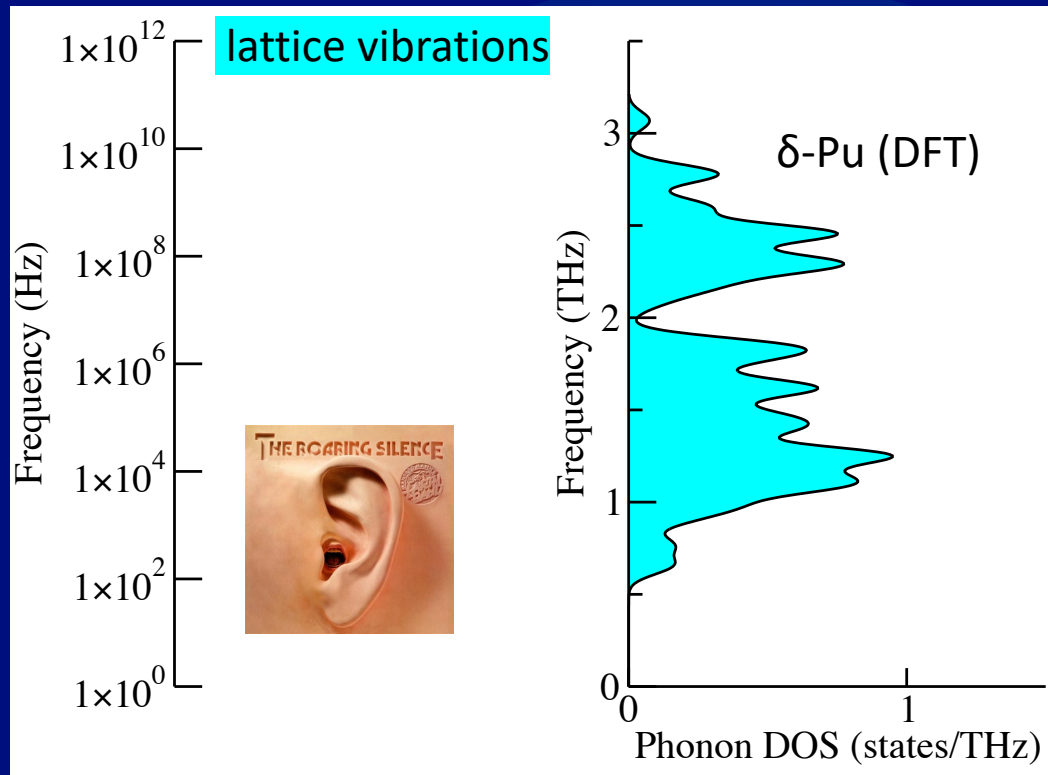
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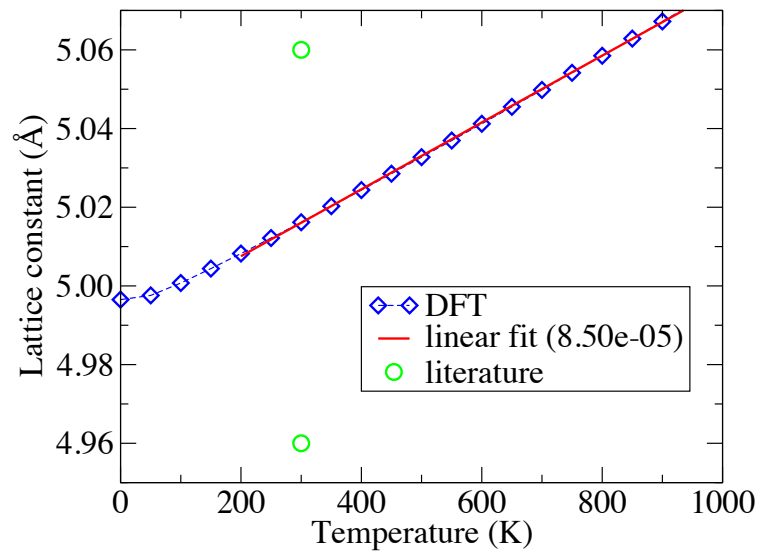
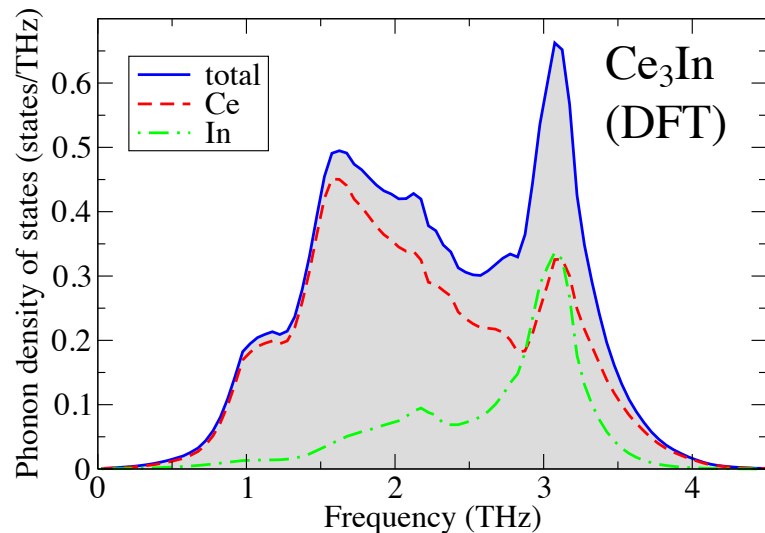
Phonons as a means for understanding Pu-Ga materials

Sven P. Rudin, T-1

December 14, 2021



DFT: Ce_3In as “cold” system to align experiment and theory – change f electron treatment?

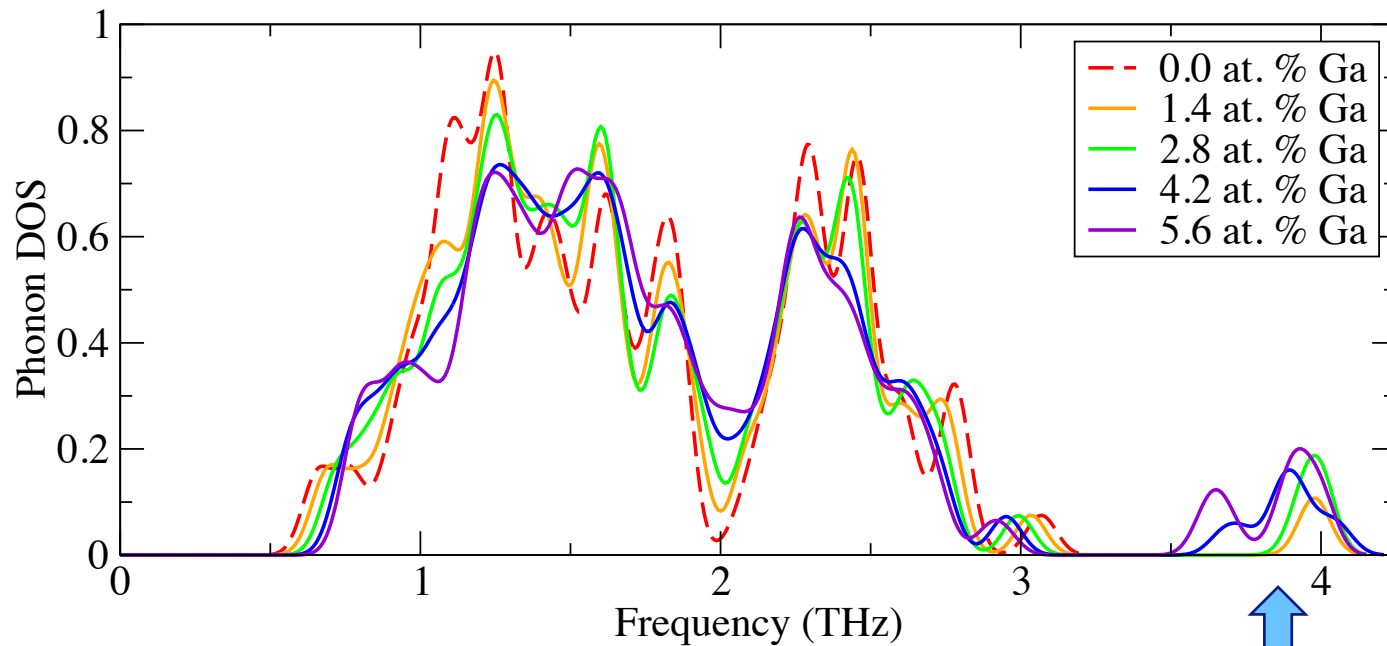


Heat capacity... previous talk:
cannot ignore electronic contribution.

Theory (LANL)	a at 300 K	5.02
	da/dT	8.5E-05
	$1/a da/dT$	1.693E-05
Experiment (LANL)	$1/a da/dT$	1.245E-05



DFT: Ga doping of δ -Pu introduces phonon modes at experimentally observed frequencies.



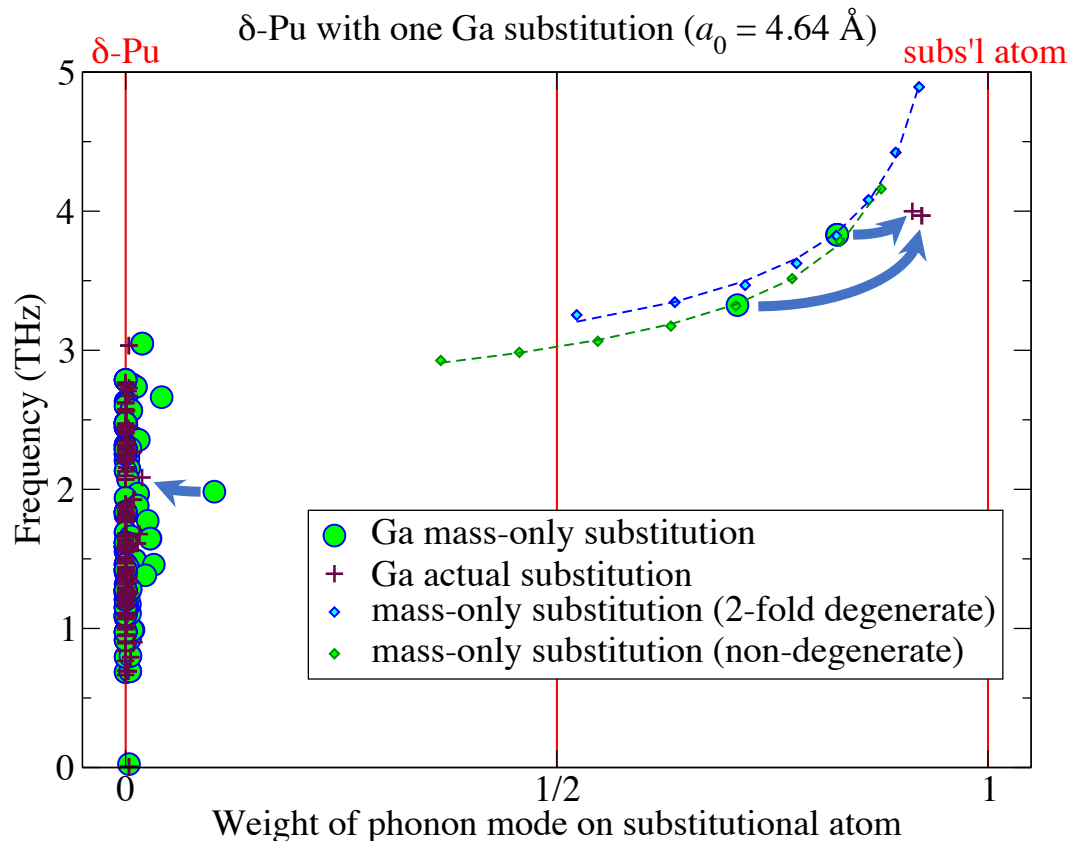
Measured for Ga in Pu-3.6 at. % Ga: $f_{\text{average}} = 3.96$ THz

LYNN et al. PRB 58, 11414 (1998)

new modes
(interacting Ga atoms at
higher concentrations)



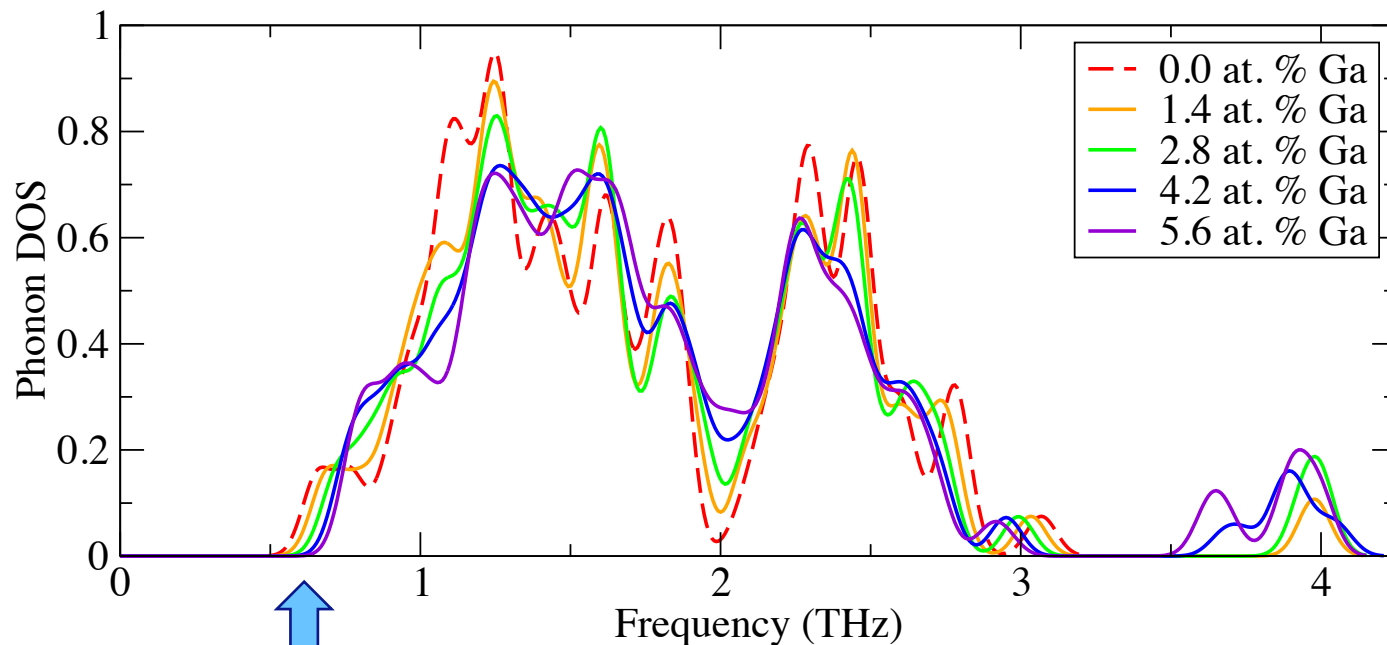
DFT: Ga doping of δ -Pu introduces phonon modes that are Ga-dominated and localized.



Compared to mass-only substitution:

- 1) Pu-dominated modes
- 2) Ga-dominated modes
 - a) more localized
 - b) higher frequencies

DFT: Ga doping of δ -Pu suggests stabilization of delta phase is not thermodynamic.



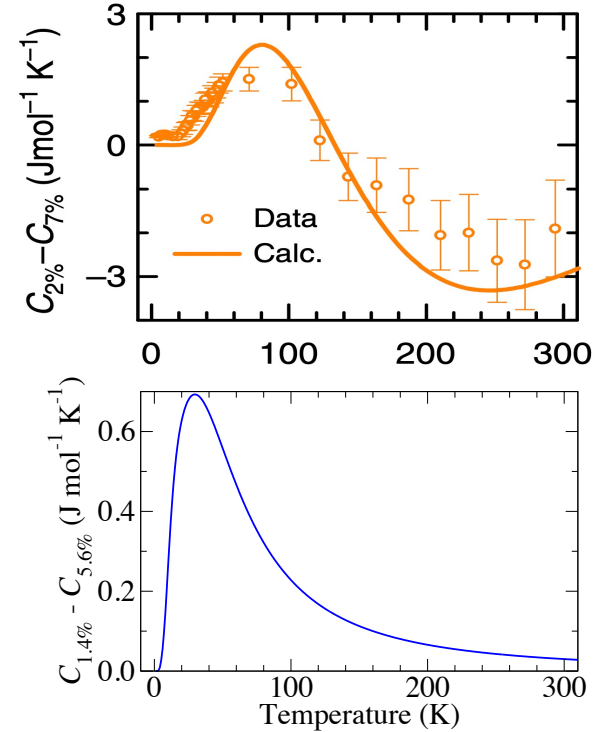
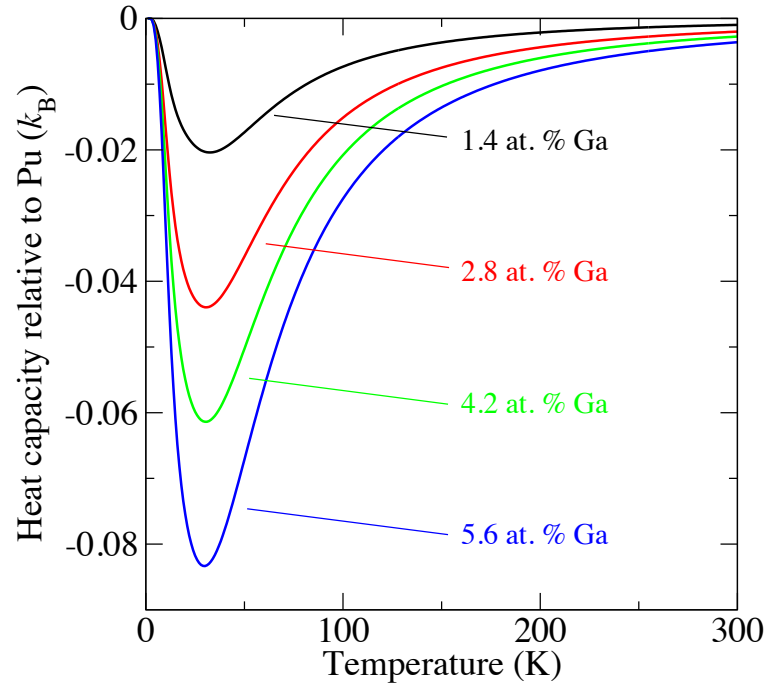
stiffening

of low-frequency modes



- thermodynamics: delta phase less stable
- proposed pathways to alpha: more stable

**DFT: Ga doping of δ -Pu changes the phonons' contribution to the heat capacity.
(But the effect is swamped by the electronic contribution.)**



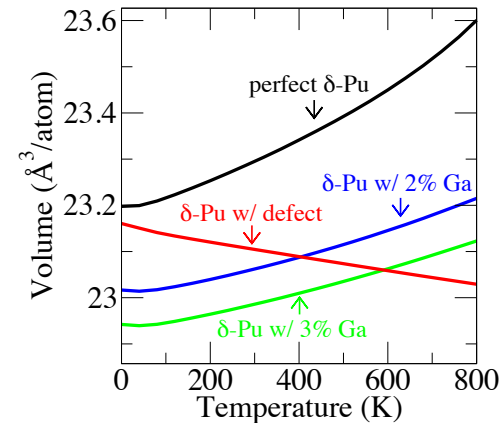
HARRISON et al.

Nat. Comm 10, 3159 (2019)



Next steps

1. Ce_3In : modify treatment of f electron in calculations.
2. DFT calculations of Ga-doped α -Pu: how does α -Pu's thermodynamic stability change?
3. Repeat the calculations with Al and In instead of Ga: do trends emerge consistent with experimental observations?
4. Explore with DFT the energy landscapes defined by the phonons proposed to initiate α -to- δ transition pathways: what do the landscapes look like? Does Ga doping change them?
5. DFT calculations of the effect of Ga doping and interstitial defect: how does Ga affect the instability induced by the defect?

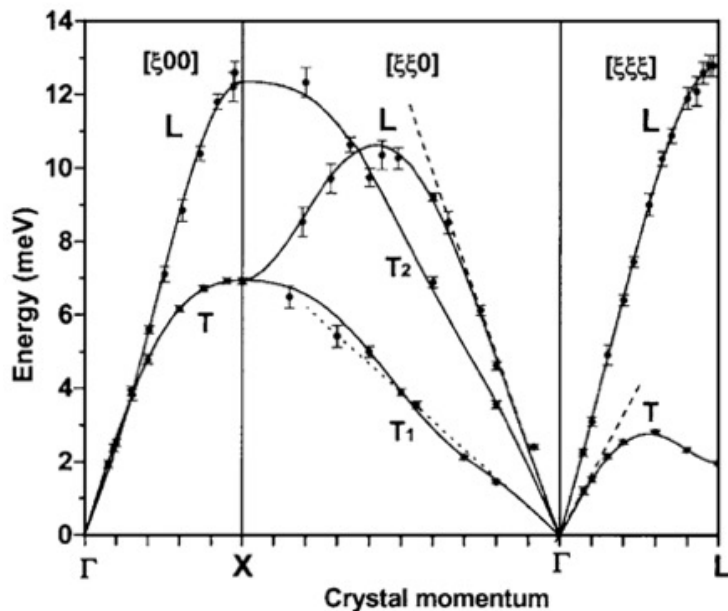


DFT: effect of magnetic structure on phonon modes

Pu-2 at. % Ga

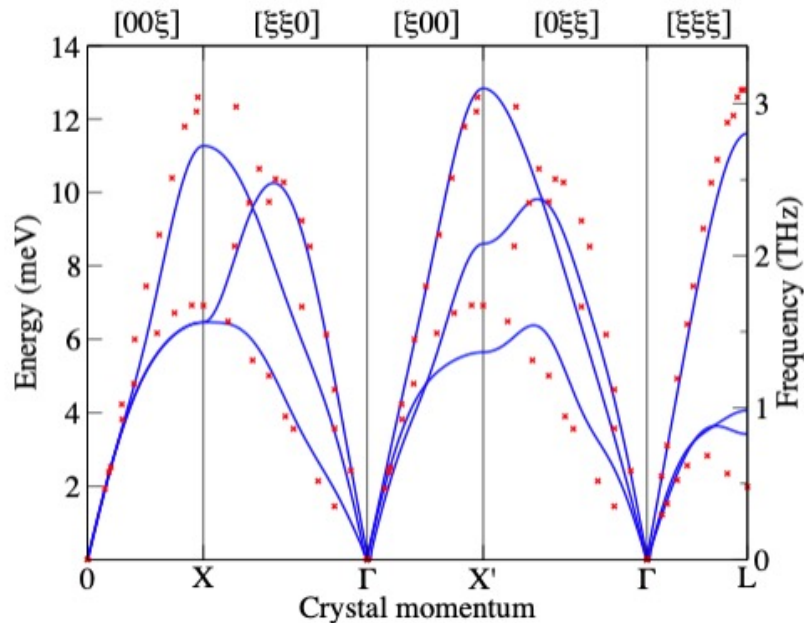
WONG *et al.* Phys Rev B 72, 064115 (2005)

Inelastic x-ray scattering

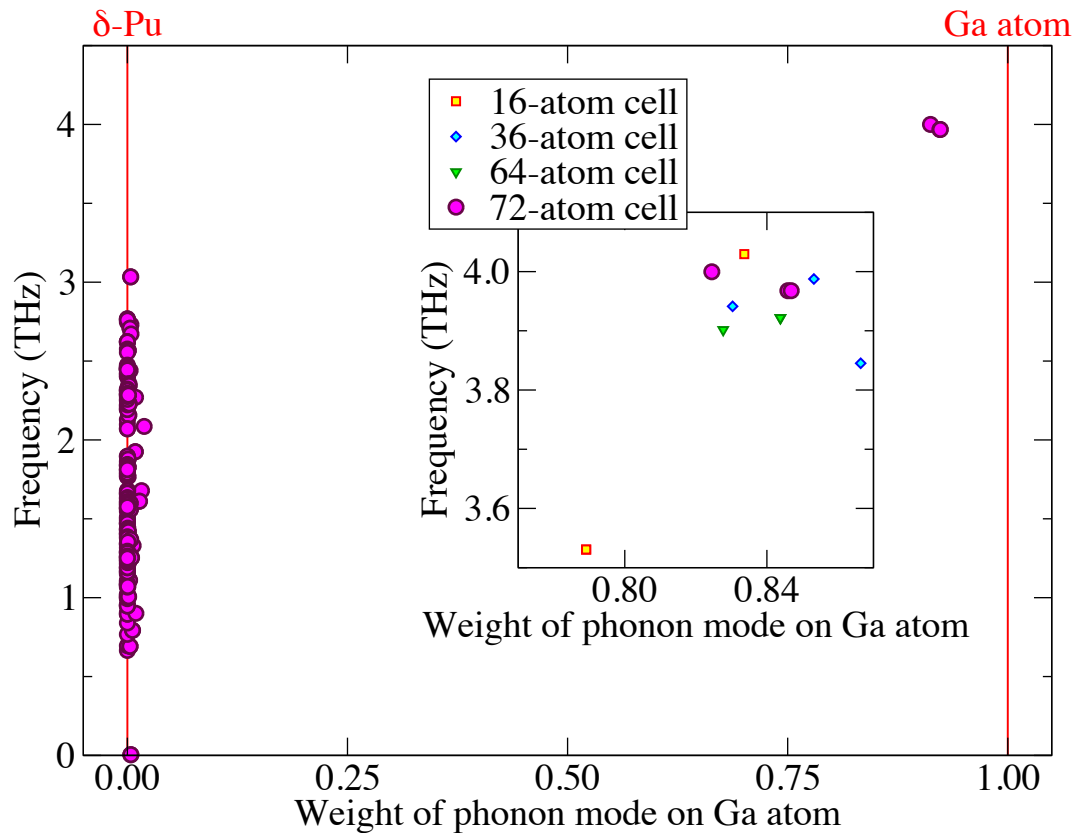


Pu-0 at. % Ga

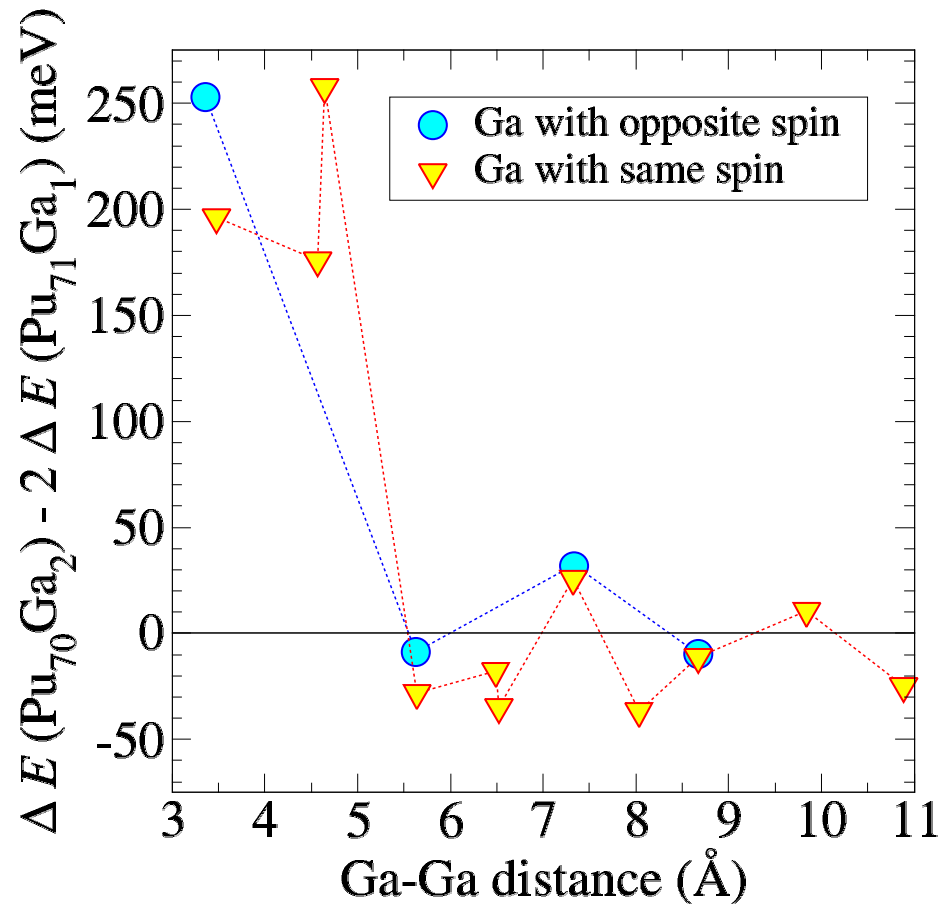
DFT (AFM)



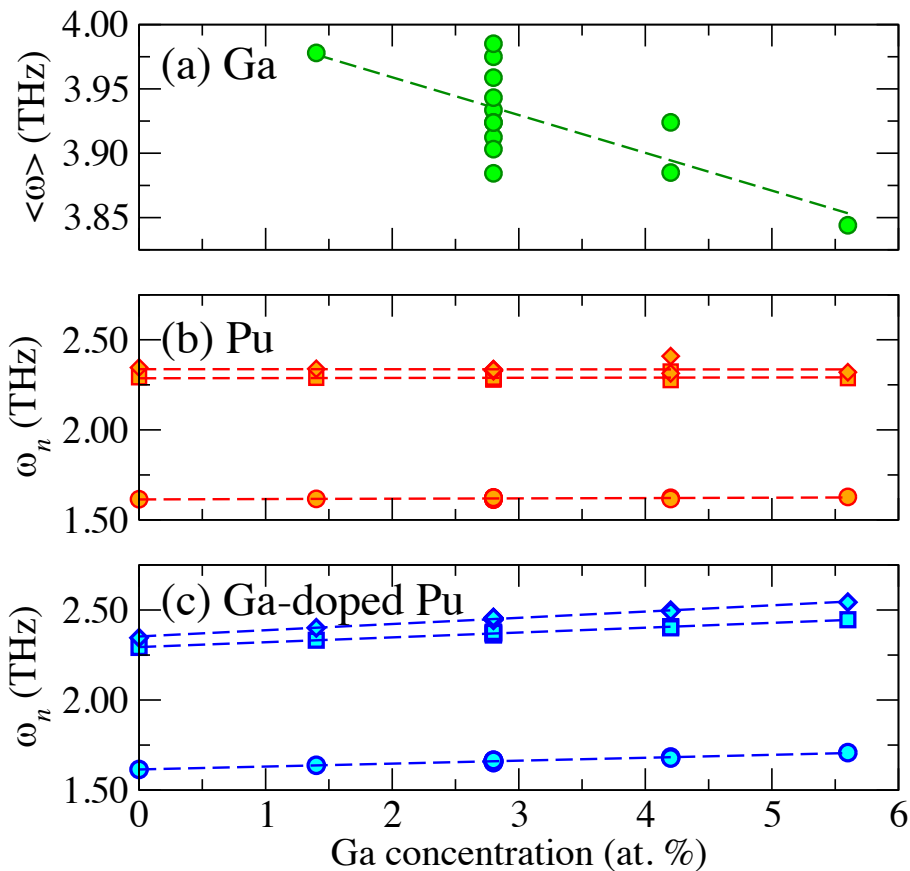
Choice of 72-atom cell size



Range of Ga-Ga cold interaction in 72-atom cell

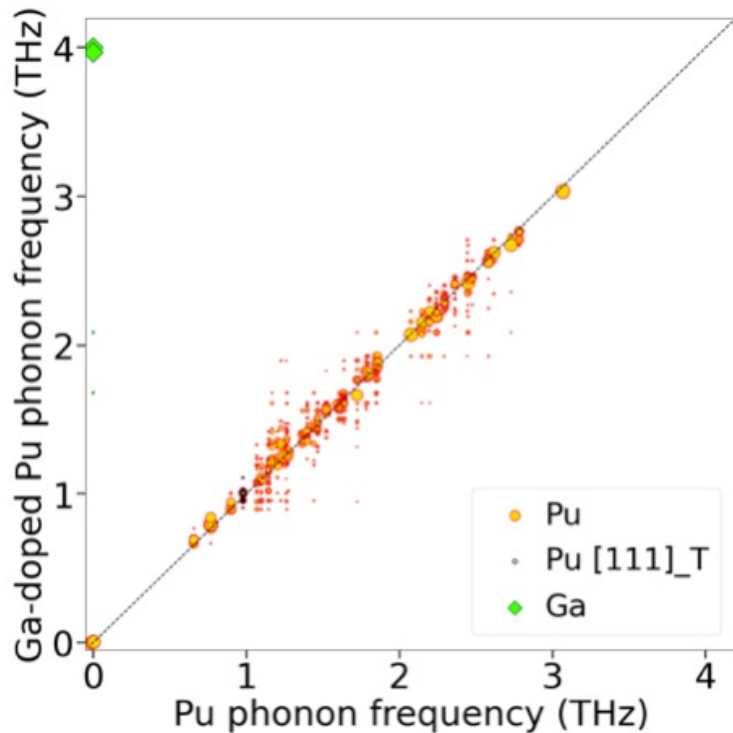


Phonon moments: change with Ga concentration



DFT: effect of Ga doping on character of low-frequency phonon modes

Pu-1.4 at. % Ga



Pu-5.6 at. % Ga

